BNWL-1115

UC-23

3 -
6-69

THE POTENTIAL OF USING
MIXED FISSION PRODUCTS
AS A SOURCE OF ENERGY

June 1969

AEC RESEARCH &
DEVELOPMENT REPORT

<i>R.E. Polto</i>	<i>38259-325</i>	<i>AUG 20 1969</i>
<i>R.W. Steiner</i>	<i>38291-703</i>	<i>MAR 4 1970</i>

BNWL-1115

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

PACIFIC NORTHWEST LABORATORY

RICHLAND, WASHINGTON

operated by

BATTELLE MEMORIAL INSTITUTE

for the

UNITED STATES ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(45-1)-1830

3 3679 00061 4802

BNWL-1115

UC-23, Radioisotope
and Radiation Applications

THE POTENTIAL OF USING MIXED FISSION PRODUCTS
AS A SOURCE OF ENERGY

By

D. W. Bolme

Chemical Technology Department
Chemistry and Metallurgy Division

June 1969

FIRST UNRESTRICTED
DISTRIBUTION MADE

JUN 27 '69

BATTELLE MEMORIAL INSTITUTE
PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON 99352

Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U.S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

THE POTENTIAL OF USING MIXED FISSION PRODUCTS
AS A SOURCE OF ENERGY

D. W. Bolme

ABSTRACT

This study estimates the amount of mixed fission products (waste byproducts obtained in the chemical reprocessing of nuclear fuels) potentially available through the year 2000, describes their probable characteristics when packaged in a form suitable for long term storage, and assesses the potential usefulness of these packages as sources of thermal and gamma (radiation) energy.

The amounts potentially available depend upon the development of the nuclear (electric) power industry. Initial availability could be as early as 1975, substantial amounts (4800 packages per year) are expected to be available in about 1990.

Packaging the waste will consist of converting the waste to a solid and depositing the solid in a metal cylinder which would then be welded shut. Typical package dimensions might be those of a 10 inch (diameter) schedule 40 stainless steel pipe 8 1/2 feet long with a filled height of 8 feet and a total weight of 1000 lb.

The characteristics of the mixed fission product cylinders as sources of gamma irradiation are roughly those of a heavily self-shielded source of cesium-134 and 137 which is contaminated with enough cerium-praseodymium-144 to cause shielding problems during transport and handling. Gamma power delivered external to a cylinder containing 2 year old waste will be about 1020 watts including 240 watts from ^{137}Cs , 585 watts from ^{134}Cs , 100 watts from $^{106}\text{Ru-Rh}$, 37 watts from $^{144}\text{Ce-Pr}$ and 58 watts from all other nuclides.

Packaged mixed fission products are not expected to be useful in mobile radiators because of the heavy shielding

made necessary by the $^{144}\text{Ce-Pr}$. Use of mixed fission products in stationary radiation facilities may be feasible for uses which require a source strength greater than that of 5 megacuries of cobalt and can tolerate large, bulky, diffuse sources.

The packaged mixed fission products (cylinders) are not economically useful sources of thermal energy except, possibly, as a source of process steam for the plant which produces the cylinders.

CONTENTS

LIST OF TABLES	vii
LIST OF FIGURES	vii
SUMMARY AND CONCLUSIONS	1
INTRODUCTION	5
Organization	6
AVAILABILITY	6
Nuclear Power Forecast	6
Mixed Fission Product Forecast	7
CHARACTERISTICS	7
Fission Product Distribution	8
Gamma Spectrum	9
Comparison of BWR and PWR Wastes	10
Packaging and Its Effects	11
Self-Shielding and Cylinder Wall Shielding	12
Fast Flux Reactor Wastes	15
ECONOMIC EVALUATION	16
Assumptions	17
Cask Description	20
Size and Type of Facility	20
Shipping and Handling Cost	22
Purchase and Maintenance of Source	23
Mixed Fission Products	23
Cesium-137 and Cobalt-60	24
Discussion	24
Possible Uses, Irradiation	26
Possible Uses, Thermal Energy	29
Value	31
Plant Size and Cost Limitations	31
Design Constraints	32
REFERENCES	37
APPENDIX A: TABLES	A-1
APPENDIX B: FIGURES	B-1

TABLES

1	Nuclear Power Forecast	A-1
2	Availability of (PWR and BWR) Mixed Fission Product Cylinders	A-2
3	Amount of Wastes in PWR and BWR Fuels Aged Two Years from Reactor Discharge	A-2
4	Gamma Spectrum of PWR Mixed Fission Products	A-3
5	Gamma Spectrum of BWR Mixed Fission Products	A-4
6	Major Sources of Gamma Power	A-5
7	Effect of Self-Shielding and Cylinder Wall Sheilding	A-5
8	Power Level of PWR and BWR Cylinders	A-6
9	Energy from One Cylinder of PWR Mixed Fission Products	A-7
10	Cost of 70 Kilowatt Gamma Source	A-8

FIGURES

1	Installed Capacity - Nuclear Electrical Generation	B-1
2	Gamma Spectra for PWR Mixed Fission Products (2 yr from reactor discharge)	B-2
3	Gamma Spectra for PWR Mixed Fission Products (3 yr from reactor discharge)	B-3
4	Gamma Spectra for PWR Mixed Fission Products (5 yr from reactor discharge)	B-4
5	Gamma Spectra for PWR Mixed Fission Products (10 yr from reactor discharge)	B-5
6	Gamma Spectra for PWR Mixed Fission Products (20 yr from reactor discharge)	B-6
7	Comparison of Gamma Spectra from PWR and BWR Wastes	B-7
8	Comparison of Mixed Fission Products and ^{137}Cs Gamma Watts	B-8

THE POTENTIAL OF USING MIXED FISSION PRODUCTS
AS A SOURCE OF ENERGY

D. W. Bolme

SUMMARY AND CONCLUSIONS

This study examines the potential usefulness of mixed fission products as a source of thermal and gamma energy. It is assumed that civilian nuclear fuel reprocessing plants will be solidifying and packaging power reactor wastes because of safety considerations and that use of these wastes (i.e., mixed fission products) as sources of energy would involve only incremental costs.

The conclusions reached in this study are that:

- The amount of solidified mixed fission products potentially available for use will become significant no sooner than about 1975.
- The packaged mixed fission products (cylinders) are not economically useful sources of heat. Even ignoring capital cost, the cost of handling the cylinders would be more than the heat available is worth. The one possible exception involves recovery of process steam from the short-term storage facility.
- Packaged mixed fission products are not expected to be useful in mobile radiators because of the heavy shielding made necessary by the $^{144}\text{Ce-Pr}$.
- Use of mixed fission products in stationary radiation facilities, particularly very large ones, does appear feasible provided the facility can be designed to accommodate these heavy, bulky, sources. Mixed fission products do not start to compete with cobalt-60 until a source equivalent to about 5 megacuries of cobalt is required.

Summarizing in more detail; if mixed fission products were available for cost of freight, roughly \$1050 to obtain one cylinder and \$600 to dispose of it 18 to 20 years later, and cobalt-60 cost 25¢ per curie delivered, a radiation facility using mixed fission products to provide 70 kilowatts of gamma power (equivalent to 4.8 megacuries of cobalt-60) could justify an additional \$1,340,000 in capital cost and \$160,000 in annual operating expense. Extra capital costs made necessary by the use of mixed fission products might typically include a 50 ton crane instead of a 5 ton crane, a 25% increase in shielding thickness, and a facility large enough to accommodate source wells a foot in diameter instead of a few inches in diameter.

The characteristics of the mixed fission product cylinders as sources of gamma irradiation are roughly those of a heavily self-shielded source of cesium-134 and 137 which is contaminated with enough cerium-praseodymium-144 to cause shielding problems during transport and handling. Gamma power delivered external to a cylinder containing 2 year old waste will be about 1020 watts including 240 watts from ^{137}Cs , 585 watts from ^{134}Cs , 100 watts from $^{106}\text{Ru-Rh}$, 37 watts from $^{144}\text{Ce-Pr}$ and 58 watts from all other nuclides.

The amount of mixed fission products potentially available is large. Tentative plans are being developed for solidifying these materials and packaging them in steel cylinders. Potential annual production is 380 cylinders in 1975, 1510 in 1980, 3070 in 1985, and 4800 in 1990. A typical cylinder might be 10 inches (nominal) diameter, 8.5 feet tall, and contain the fission products produced by approximately 48,000 megawatt days of fuel exposure.

It will probably be advantageous to hold these cylinders in a short-term cooling facility at the site of production until

the wastes are 2 years out of the reactor. Use of these cylinders as a source of thermal heat does not appear attractive except, possibly, while they are in this short-term cooling facility. Such a facility, receiving 1000 cylinders per year and storing them 1 1/2 years, might justify an (additional) capital cost of \$300,000 to \$500,000 for the modifications necessary to produce process steam.

Major amounts of fast flux reactor wastes are not expected to become available until 1990 at which time they might account for 20% of the wastes available. Except for greater amounts of ^{106}Ru the fission product distribution of fast reactor wastes will be roughly the same as BWR or PWR wastes. Because of the extra ruthenium, cylinders containing these wastes might require longer "short-term storage" than cylinders of BWR or PWR wastes but could then be handled in much the same manner.

Cesium separated from power reactor waste is a possible competitor. The sources, if packaged as a part of the waste solidification program might typically be 2-inch diameter rods 2 feet long containing 60 kilocuries. They would be cheaper to transport and use. The relative economics of mixed fission products versus separated cesium would depend strongly on how the cost of separation was apportioned between the user of the radiation source and the operator of the separations plant (who might separate the Cs [and Sr] in any case in order to make the long term storage of wastes easier).

Separated strontium-90 may also be useful but primarily as a source of heat. Since the mixed fission products are not useful as a source of heat, competition between mixed fission products and strontium-90 does not exist.

INTRODUCTION

This report presents information on the amount of mixed fission products which is expected to be available from the nuclear power industry, their characteristics (thermal watts and gamma spectrum as a function of time) and an estimate of the cost of using these materials in car-lot quantities.

The growth predicted for nuclear power in electric generation will result in increased production of (by-product) fission products. These fission products, separated as a waste when the spent fuels are processed to recover uranium, must be prevented from entering the environment. One method of accomplishing this is to convert these materials to a solid form, suitably packaged, and hold them in very long-term storage. The entire cost of such an operation would have to be borne as a cost by some part of the national economy unless profitable use can be found for the fission products.

A substantial amount of work has already gone into investigations of the use of specific isotopes which can be separated from the by-product fission products. This work continues. The separated materials are fairly expensive, however, and the apparent market is not large enough to use all of the radioactive material which will be available. Additional uses, perhaps large scale, may exist if the radioactivity present can be made available in a less expensive form. One possibility is to use the crude mixture of fission products solidified and packaged in the same form as required for long-term (non-use) storage.

This report presents the results of a preliminary study undertaken to estimate 1) the amount of mixed fission products potentially available, 2) the characteristics expected of

packaged crude fission product mixtures, and 3) the (apparent minimum) cost which might be incurred in using these materials in car-lot quantities. A brief discussion of possible methods of using the material is included.

ORGANIZATION

This report is, among other things, a summary of a large amount of detailed data. To allow easy perusal of the data, tables and figures are contained on foldout sheets in Appendices A and B, respectively.

AVAILABILITY

NUCLEAR POWER FORECAST

The amount of mixed fission products which will be available is dependent upon the growth in the use of nuclear powered electrical generating plants. The growth expected of this industry has been predicted by a number of people and organizations. One recent, thorough study is that made as part of an isotope production and availability report for the Atomic Energy Commission by McKee, et al. (1968). The growth rate they predicted for the "Most Likely Combination of Liquid Water Reactors Recycling Plutonium and Uranium (U-236) with Introduction of Fast Reactors in 1980" is presented in the first six columns of Table 1 and in Figure 1. The amount of cesium-137 in the spent fuel discharged from the reactors is presented as a function of time in Column 7 of Table 1. (These figures are McKee's but differ from his "Annual Availability of Fission Products," because they do not include a 10% loss in processing, a 2 year delay between "discharge" and "available" and the radioactive decay which would occur during this period.)

MIXED FISSION PRODUCT FORECAST

The tonne (metric tons) of spent fuel discharged each year by each type of reactor can be calculated from the data just presented.* This has been done and the results are presented in the last three columns of Table 1.

As explained later, this study assumes that the wastes from the fuels will be solidified and packaged in cylinders containing, alternatively, the wastes from 1 1/2 tonne of PWR fuel or 2 tonne of BWR fuel, and that these cylinders will be stored until the wastes are 2 years out of the reactor before they are available. Under these conditions, the rate at which cylinders become available will be that presented in Table 2.

CHARACTERISTICS

The characteristics of the mixed fission products potentially available are those of the fission products themselves

* Details of fission product distributions and calculations are presented later. For the purposes of this study, PWR fuels were assumed to have a uniform exposure level of 30,000 MWd/t and BWR fuels 25,000 MWd/t. The amount of ^{137}Cs discharged per megawatt day of exposure was almost identical [PWR was 1.005(BWR)]. If it is assumed that the overall thermal efficiency of the PWR and BWR are the same, and this appears probable, the amount of ^{137}Cs produced per unit of electricity becomes independent of the type of reactor. Knowing the total ^{137}Cs produced, the fraction of power produced by each reactor and the amount of ^{137}Cs per tonne of spent fuel, the tonne (of each type) of fuel to be discharged each year can be calculated. The same approach may be used in predicting fast flux reactor (spent) fuel tonnage. The fraction of fissions which produce ^{137}Cs is slightly higher in a fast reactor (about a 5% increase) but the power produced per fission is also increased (about 7% less neutrino production) so the amount of power per unit of ^{137}Cs will remain about the same, about 26,400 MWd per kg ^{137}Cs .

as modified by the matrix in which they are contained, the geometry and shielding characteristics of the package, and age.

FISSION PRODUCT DISTRIBUTION

The fission product distributions used in this study were predicted by the use of ISOGEN (a computer program originally designed to predict the fission product distributions produced by the Atomic Energy Commission's production reactors) which was modified to use power reactor cross sections [Van Tuyl (1964)].

This program uses a "chain tracing" technique to calculate the amount of each daughter produced from each of the initial parents. In doing this, the program checks each step and, if the first (Bateman equation) sequence is not suitable, selects one of three algebraic rearrangements of the Bateman equation so as to maintain a numerical accuracy of at least 0.1% (which is as much as the data normally warrant). Design of the program is such that it can accommodate any actual system of nuclides including branching, neutron capture, fission and feedback at any or all nuclides. As used in this study, the program calculated amounts of all nuclides between ^{72}Zr and ^{166}Dy having a half life greater than one day, all daughters of such nuclides, and major nuclides between ^{203}TI and ^{250}Cm . It can also be used, in a similar fashion, for half lives greater than 1 hour or greater than 10 minutes.

The conditions used in predicting the PWR and BWR fission product distributions are as follows:

<u>Initial Fuel</u>	<u>PWR</u>	<u>BWR</u>
^{235}U	33.0 kg	25.6 kg
^{238}U	967.0 kg	974.4 kg
<u>Operating "Year"</u>		
Operating	105 days	105 days
Down	14 days	14 days
Operating	105 days	105 days
Down	14 days	14 days
Operating	98 days	98 days
Down	28 days	28 days
Number of years	3	4
Flux	2.2×10^{13}	1.54×10^{13}
<u>Number of fissions</u>		
^{235}U	4.655×10^{25}	3.520×10^{25}
^{239}Pu	2.104×10^{25}	1.951×10^{25}
^{238}U	1.399×10^{25}	1.345×10^{25}
240+	3.696×10^{22}	2.529×10^{22}
Total	8.161×10^{25}	6.819×10^{25}
Indicated Exposure	30,000 MWd/tonne	25,000 MWd/tonne
Power Level	32.5 MW/tonne	20.3 MW/tonne

The computer program actually calculated the amount of each of 300 nuclides expected to exist in the fuel. Those nuclides of major importance and their amount in wastes 2 years out of the reactor are presented in Table 3. These data are based upon all nuclides in the discharged fuel less all curium, americium, plutonium, and uranium (which would be separated for recycle) and krypton (which would not be included in the solidified waste).

GAMMA SPECTRUM

The gamma spectrum emitted by the radionuclides obtained from 1 tonne of each of the two fuels is presented as a function

of time in Tables 4 (PWR) and 5 (BWR). The γ power (watts) identified with a particular γ energy (MeV) are those associated with the nuclide shown. Those not so identified are a summation of all the γ power (watts) contributed at all energy levels falling between the two adjacent designated levels.

In developing Tables 4 and 5, only major contributors of energy were considered. The individual nuclides were ^{155}Eu , ^{154}Eu , $^{148\text{m}}\text{Pm}$, ^{147}Pm , ^{144}Pr , ^{144}Ce , ^{141}Ce , ^{137}Cs , ^{134}Cs , ^{129}Te , $^{129\text{m}}\text{Te}$, ^{127}Te , $^{125\text{m}}\text{Te}$, ^{125}Sb , $^{110\text{m}}\text{Ag}$, $^{103\text{m}}\text{Rh}$, ^{106}Ru , ^{103}Ru , ^{95}Nb , $^{95\text{m}}\text{Nb}$, ^{95}Zr , ^{91}Y , ^{90}Y , ^{90}Sr , ^{89}Sr . The effect of omitting the other nuclides is not important. For example, with PWR fuel at 0.5 years from reactor discharge the nuclides considered accounted for 4620 out of 4623 gamma watts present with the biggest single source of error the exclusion of 1 watt from the La daughter of 12.8 day ^{140}Ba .

The gamma spectra for PWR fuels is presented graphically in Figures 2 through 6 at ages of 2, 3, 5, 19 and 20 years. In these figures the solid vertical lines represent the gamma power (watts) released at the specific energy level indicated. The small circles represent the gamma power released between the energy levels which involve major amounts of power.

COMPARISON OF BWR AND PWR WASTES

The distribution of gamma power from BWR wastes is very similar to that from PWR wastes. Detailed examination of Tables 4 and 5 will indicate this; however, it may be inferred more easily from Table 6 which compares the fraction of total gamma power (watts) contributed by individual nuclides in each case. Less than 5% of the total gamma power originates in nuclides not common to both materials.

Alternatively, the power levels of the major peaks emitted by PWR and BWR materials at 2 years from discharge are compared in Figure 7. As may be seen, the two materials are quite similar in gamma ray spectra. This similarity increases with age as the short-lived materials decay and ^{137}Cs becomes the sole important source of gamma power in both cases.

PACKAGING AND ITS EFFECTS

The package assumed in this study is a 10 inch (nominal) diameter schedule 40 stainless steel pipe about 8.5 feet long with a filled height of 8 feet. Actual dimensions would be 10.75 inch OD; 10.02 inch ID, 0.365 inch wall thickness, and the volume of the mixed fission products would be 4.38 cubic feet. The total package weight would be about 1000 pounds. The steel cylinder can routinely be heated during the filling operation to temperatures high enough to cause heat treatment of the steel and produce a surface having a thermal emissivity of about 0.85. Under normal conditions, the cylinders will be able to discharge all of their internally generated heat by radiation if this is advantageous. Current designs in the Waste Solidification Program require that the external temperature of the cylinder be held below 425 °C but at this temperature it could radiate heat at a maximum rate of 3630 Btu/hr (1.06kW) per square foot or, neglecting heat loss from ends, 81,700 Btu/hr (23.9 kW) from the cylinder (less reflection and radiation from the surroundings). This may be compared with the total energy release, of the cylinder, of about 6.6 kW/cylinder for 2 year wastes.

It has previously been stated that each cylinder would contain the by-product or waste fission products from 1.5 tonne of PWR fuel or 2.0 tonne of BWR fuel or their equivalent. A number of considerations enter into the choice of cylinder

diameter; filled height, and loading density, the most restrictive of which involves the removal of heat when the cylinder contains 0.5 year waste (i.e., at time of filling). The configuration chosen and described above is obtainable with present technology.

Self-Shielding and Cylinder Wall Shielding

The extent of self-shielding is dependent upon the density and composition of the matrix in which the fission products are contained. Precise calculation of self-shielding is therefore not possible until a particular waste solidification process is chosen for use. A pilot plant investigation of three alternative processes (a phosphate glass process, a pot calcination process, and a spray calcination process) is presently being conducted under the auspices of the Atomic Energy Commission.

An approximation of the self-shielding is reported below in terms of watts delivered at the outside of the package. These calculations are based upon the assumption that the self-shielding characteristics of the solidified mixed fission products will be the same as an equivalent volume of normal concrete with a specific gravity of 2.3. In addition to self-shielding, the effect of the container wall, which will provide shielding equivalent to 0.365 inches of iron, has been included.

Gamma Rays with Initial Energies Less than 0.5 MeV will make little contribution to energies delivered external to the package. The power (watts) contributed by 0.00 to 0.14 MeV gammas are about 1.5% of the total gamma power released by the mixed fission products and less than one-tenth of this will penetrate to the exterior of the cask. The watts contributed by 0.14 to 0.50 MeV gammas are less than 3/4% of the total gamma power emitted and less than

one-fifth of this is expected to appear as gamma power available at the surface of the cylinder.

Gamma Rays with Initial Energies Between 0.5 and 0.8 MeV contribute about 90% of the total gamma watts released. The weighted average absorption coefficient for this series of gamma emissions is substantially identical to that of the 0.662 MeV gamma from ^{137}Cs . Standard calculations (Rockwell, Reactor Shielding Design Manual) indicate that the self-shielding expected for 0.66 MeV gamma source, uniformly imbedded in a 10 inch cylinder of concrete, would be the same as that of a line source shielded by 2.5 inches of concrete. Under these conditions, the attenuation of the primary would be 0.3166 in the concrete (self-shielding) and 0.59 in the cylinder wall for a total primary attenuation of 0.187. The buildup factor (compensation for forward scattering) is more difficult to obtain. It would have to be greater than 1.62 and less than 3.35 with the most probable value between 2.5 and 3.0. For purposes of this calculation, 2.8 which is what would be predicted for an iron shield producing a primary attenuation of 0.187) will be assumed so that total external gamma watts would be $(0.187)(2.8) = 0.52$ or 52% of that released.

Gamma Rays with Initial Energies above 0.8 MeV contribute less than 8% of the total gamma power released by the mixed fission products. The majority of this is centered about 1.25 MeV and would have a primary attenuation of about 0.3 and a buildup factor of about 2.5. The $^{144}\text{Ce-Pr}$ gamma at 2.18 MeV would have a primary attenuation of about 0.4 and a buildup factor of about 2.0.

Bremsstrahlung will not make a significant contribution to the gamma rays emitted from the packaged wastes. An example may make this obvious. The beta rays from the ^{90}Sr -Y associated with 1 kilocurie of ^{90}Sr will produce about 1 curie of 0.6 ± 0.05 MeV gamma rays. When first filled with the 2-year-old wastes from 1.5 tonne of PWR fuel, one cylinder would contain 103.5 kilocuries ^{90}Sr which would emit 103.5 curies of 0.6 ± 0.05 MeV gamma rays, equivalent to about 0.37 gamma watts which is small compared to the total emission of 1283 gamma watts. Yields are higher at lower energies, but these would be removed by self-shielding and cylinder wall shielding; at higher energies the yields are too low to be important as far as gamma power is concerned. A similar argument holds true for shielding considerations; the amount of 2.0+ MeV gammas contributed by ^{90}Sr -Y bremsstrahlung is inconsequential in comparison to the primary gammas of ^{144}Ce -Pr.

The total of these effects would be such that the fraction of the gamma power (watts) released which would be delivered external to the cask would be, roughly, 53% as shown in Table 7.

Based upon these estimates the thermal and gamma power delivered exterior to cylinders of PWR and BWR wastes are presented, as a function of time, in Table 8.

Figure 8 presents a comparison between the gamma power delivered exterior to PWR and BWR cylinders and that from 100,000 curies of cesium-137. It should be noted that the gamma spectrum of the cesium-137 will be almost entirely 0.662 MeV γ , while almost two-thirds of the energy from the mixed fission products will be the result of buildup and occupy all energies between ~ 0.15 MeV and 0.8 MeV.

One of the problems of mixed fission products is immediately apparent. Even if the material is held in "short term cooling" until 2 years out of the reactor, it still has a high initial decay rate. In the end analysis it is the integrated dose (and therefore the integrated power) which is purchased. The approximate kilowatt hours, both gamma and thermal, which will be delivered each year by one cylinder of PWR waste are presented as a function of time in Table 9.

In summary, the packaged mixed fission products may be characterized as large sources of ^{134}Cs and ^{137}Cs gamma rays which have 1) self and package shielding sufficient that gamma power delivered is 53% of that generated and 2) enough $^{144}\text{Ce-Pr}$ and ^{106}Ru to make shielding for protection during transport etc., a major problem.

FAST FLUX REACTOR WASTES

The amount of power produced in fast reactors is not expected to become substantial until about 1985. Since there are delays between power production and fuel discharge and between fuel discharge and fuel processing, the wastes from fast reactors are not expected to be important until about 1990 at which time amount of fast reactor wastes would be in approximate proportion to the power production in 1985, roughly, 20%.

The distribution of major fission products in the waste of the fast reactors, particularly after a few years of cooling, is controlled more by the identity of the nuclide fissioned than by the spectrum of the flux. The major difference in the fast reactor waste will arise in the greater importance of plutonium as a source of fission products. The only major radionuclide this affects is ^{106}Ru , and a ratio of $^{106}\text{Ru} : ^{137}\text{Cs}$ in LMFBR wastes

will be 3.5 to 4.0 times than in PWR wastes. These wastes would probably be packed at the same "density" (MWd of fission products per cubic feet of waste) as PWR and BWR wastes. They might require longer storage before release for use. After such storage, however, the handling and gamma radiation characteristics of these packaged wastes would be quite similar to those of PWR or BWR wastes.

ECONOMIC EVALUATION

The economic boundaries within which mixed fission products would have to compete are not well defined. Major potential competitors are cobalt-60 produced by irradiation of the metal or cesium-137 recovered from mixed fission products; however, the price of these materials is not firmly established. Compounding these uncertainties are the questions of type of use, size of source required, and type of facility. This section

- (1) characterizes the type and size of applications for which mixed fission products have the best chance of being competitive,
- (2) estimates the cost of obtaining and maintaining a mixed fission product source of minimum economic size,
- (3) compares this capital and operating cost with those of a similar facility furnishing the same gamma watts by use of cobalt-60 and cesium-137, and
- (4) determines the difference in the costs which is, then, the additional capital and operating cost which could be justified for a facility using mixed fission products instead of cobalt or cesium.

ASSUMPTIONS

If the nuclear power industry develops as predicted, the production of mixed fission products will exceed present requirements for sources of radiation by orders of magnitude. Since the mixed fission products are a by-product they will be produced whether they are used or not. They will, therefore, be a glut on the market at least in the near term. Any mixed fission products which cannot be used will have to be stored permanently at some cost. Therefore, any use which provides "free" storage of the material would be an improvement over the alternative even if there is no payment for use of the material.

Use of mixed fission products would seem to involve competition with cobalt-60 produced by private industry. Actually this competition is more apparent than real. Cobalt can be used in facilities of any size including mobile irradiators. The largest existing facilities are in the 100 to 500 kilocurie range while there is at least one (AECL) 1.5 megacurie facility planned and one (RAMCO) facility designed for 5 megacuries under construction. In contrast, it will be shown that mixed fission products cannot be used in mobile irradiators and the smallest potentially economic facility would be in the 1.5 to 5 megacurie range.

Major assumptions, their rationale, and their effect upon the estimated cost are as follows:

- (1) It is assumed that solidified mixed fission products, in suitable containers, will be made available to the user at zero cost FOB the short term cooling facility and will be accepted from him at no cost FOB the long term storage facility.

Rationale - Solidification, packaging, short term cooling, and long term storage of the mixed fission products will be necessary whether they are used or not. Since use of mixed fission products does not impose an added cost in these areas, no costs are included in the cost of using the material.

Effect - The user will pay for all extra costs involved in the use of mixed fission products; the source of the mixed fission products will be relieved of the cost of storing the mixed fission products for as long as they are used, and the cost of transporting the MFP from short-term storage facilities (presumably located at the chemical processing plant) to the long term storage facilities.

- (2) It is assumed that the mixed fission products will be packaged in one standard package which will be a 10 inch diameter schedule 40 pipe about 8 1/2 feet tall with a filled height of 8 feet and a total weight of about 1000 pounds.

Rationale - The major problem in packaging these wastes is a combination of heat removal and mechanical strength when they are first filled. The cylinder meets these problems best and appears to be the package which will be used.

Effect - No major effect except that the one standard geometry simplifies handling and transport problems.

- (3) It is assumed that all shipments of solidified mixed fission products will be made in special casks designed solely for that purpose and that the casks will be in substantially continuous use.

Rationale - The cylinders of solidified mixed fission products will have to be shipped from their point of production to permanent, long-term storage whether they are used enroute or not. The amount expected to

be shipped (see Table 2) will justify the use of special shipping casks designed to take advantage of the comparatively low cooling requirements and lack of criticality problems which shipment of solidified mixed fission products will involve.

Effect - The cost of shipping mixed fission products to use will be reduced by allowing the use of specially designed casks without imposing penalties because the casks are not completely utilized.

- (4) It is assumed that the user of the MFP will pay for use of the cask only while it is used to transport materials to his facility or from his facility to the long term storage sites.

Rationale - Same as above.

Effect - Little effect except that shared use allows the interest, depreciation, and maintenance on the cask to be charged at a constant rate per trip without having to worry as to whether the industrial use will be large enough to justify a special cask.

- (5) It is assumed that the facility using the mixed fission products will be large enough that the amount of mixed fission products required in any one shipment will be equal to all of the material that can be transported by one such cask.

Rationale - The amount of material available will be substantial. Any use large enough to result in important savings (by temporary storage of the mixed fission products) will require major amounts of the material.

Effect - The cost, per unit of material, for loading, transporting, and unloading the mixed fission products will be minimized.

CASK DESCRIPTION

The cask used in this estimate is a simple pot and lid assembly capable of containing three cylinders of solidified mixed fission products. Dimensions are:

ID - ~24.5 inches

OD - ~49.5 inches

Inside length - 8 1/2 feet

Outside length - 10 feet 6 inches

Shielding - 11 inches of lead

Weight - 73,000 pounds

Cost @ \$1/lb - \$73,000

Heat generated by the cylinders would be transferred to the inner surface of the cask primarily by radiation, through the cask wall by conduction, and from the cask wall to air by conduction and natural convection. Fins to promote heat transfer from the cask exterior to the air might be advantageous but calculations indicate they are not necessary. The cask would have to be housed during transit to meet personnel protection requirements, both radiation (no approach within 2 feet) and thermal burns from the cask exterior, and the housing would have to be louvered to allow escape of heated air.

SIZE AND TYPE OF FACILITY

Mobile facilities do not appear to offer much chance for economic use of mixed fission products. The smallest unit of mixed fission products will be one cylinder which, containing wastes 2 years out of the reactor, would supply 1000 watts of gamma power and require a 20 ton cask [to shield against the $^{144}\text{Ce-Pr}$ (and ^{106}Ru) gamma rays]. The same gamma power (watts) could be obtained from 310,000 curies of ^{137}Cs or 70,000 curies of ^{60}Co with far less weight and volume (both source and cask),

longer source life, and more flexibility in the design of the facility. Exceptions may occur, of course, i.e., a 20 year old cylinder may prove ideal for some special purpose, but mobile irradiation facilities will not offer an important use for mixed fission products.

Stationary facilities, particularly large, underground stationary facilities, appear to offer the best opportunity for use of mixed fission products. Shielding will remain a problem, of course, but an underground facility can use concrete and earth (instead of lead) and penalties for shielding weight are comparatively minor. Finally, a facility large enough to receive replacement sources in truck load lots (one truckload equals 3 cylinders in one cask weighing about 40 tons loaded) would have an advantage over one which could not. The shipment of one cylinder in a 20 ton cask instead of 3 cylinders in a 40 ton cask would cost at least 50% more per cylinder. Since shipping cost is the major cost involved in the use of mixed fission products, this is important.

The minimum sized facility capable of accepting 3 cylinder shipments for source maintenance would probably be one requiring 3 cylinders per year. Assuming cylinders were held in use for 18 years this would mean an inventory of 54 cylinders and jump in power (gamma watts) of 20% when the cylinders were replaced. A more reasonable minimum sized facility would be one requiring four 3-cylinder shipments per year for maintenance of source.

The facility upon which this estimate is based is an irradiation facility furnishing an average gamma power of 70 kilowatts. If this power were furnished by mixed fission products the source inventory would eventually amount to 216 cylinders and require replacement of 12 cylinders per year (assuming that the cylinders would be acquired at age = 2 years

from reactor and shipped to permanent storage at age = 20 years from reactor). An equivalent facility using ^{137}Cs would require an inventory of 24,000,000 curies and addition of 550,000 curies per year for maintenance of source. Use of cobalt-60 would require 5,700,000 curies and 705,000 curies per year.

SHIPPING AND HANDLING COST

The estimated cost of placing the mixed fission products into use is as follows:

Transport

Basis: One truck carrying 1 cask which contains 3 cylinders of mixed fission products.
(or one 75 ton railroad car carrying 2 casks)
(or one 150 ton railroad car carrying 4 casks)

Cost Per Cask of 3 Cylinders

Loading (furnished FOB short term cooling facility)	\$ -0-
Transport	
Shipping and return of empty cask @ \$2.50/cwt.	2000
Cask (capital cost \$73,000)	
Depreciation @ 0.5%/trip	365
Interest @ 0.2%/trip	146
Maintenance @ \$30/trip	30
Unloading	350
Transfer (3 cylinders) from unloading basin to point of use	<u>200</u>
Total	\$3091/cask \$1030/cylinder

Once the cylinders were in place it would be economic to retain them in use for at least 20 years. Eventually, however, the power level would decrease to the point where it would be

profitable to replace the old cylinders with fresh. The old cylinders would have to be transferred to permanent storage and this cost would be:

Transfer from point of use to unloading basin	\$ 200
Load into emptied cask	350
Transport to final, long term storage*	1000
Cask depreciation, interest, maintenance*	270
Unloading (furnished by long term storage facility)	<u>-0-</u>
Total	\$1820
or	\$610/cylinder

PURCHASE AND MAINTENANCE OF SOURCE

Mixed Fission Products

Purchase and maintenance of a 70 kW gamma irradiation facility using mixed fission products would involve:

1. An initial inventory of about 84 cylinders at \$1030/cylinder (enough to provide 70 kW of gamma energy at the time the first cylinders are added).
2. A periodic addition of 3 cylinders (about 9 cylinders per year) until a total of 216 cylinders was obtained in about 15 years.
3. Replacement of 12 cylinders per year at \$1640/cylinder.

Original investment would be:

84 cylinders at \$1030 per cylinder \$86,520

Source maintenance would require:

During the 1st 15 years (approximately)

Addition of 9 cylinders/year at \$1030/cylinder \$9270/yr

Beyond 15 years

The replacement of 12 cylinders/year at
\$1640 per cylinder

\$19,680/yr

* At 50% of original shipping cost because this shipment would partially replace the return of an empty cask.

In summary, provision of a source using mixed fission products to produce 70 kilowatts of gamma energy would require an initial capital investment of \$87,000 and an annual cost not exceeding \$20,000 per year.

Cesium-137 and Cobalt-60

Provision of the same 70 kilowatts of gamma energy with cesium-137 would require purchase of 24,000,000 curies initially (301 curies per gamma watt and allowing for the first year's decay and for shielding losses of 10%) and 550,000 curies per year to compensate for annual decay. Cobalt-60 would require 5,700,000 curies plus 705,000 curies per year (same basis except 5% shielding losses).

The cost of obtaining and maintaining a 70 kilowatt source of gamma energy may therefore be compared as in Table 10. The difference in source purchase and maintenance costs are the extra capital and operating cost which a facility using mixed fission products might justify. As an example, if the competitive source were cobalt-60 at 25¢ per curie, delivered, a facility of this size using mixed fission products could justify an additional capital investment of about \$1,338,000 and additional operating cost of about \$160,000 per year.

Discussion

A look at the difficulties is now in order.

The question of shipping costs for cobalt and cesium has been avoided by specifying delivered costs for cobalt-60 and cesium-137; however, the cask used to ship 3 cylinders (capable of delivering, as mixed fission products, about 3000 watts of gamma power) could, because of lower gamma energies, be used to ship about 40,000 gamma watts of cobalt-60.

The cask used to transfer individual cylinders of mixed fission products from the unloading basin to the point of use would involve 10 inches of lead, weigh 18 tons and cost \$10,000, while a cask to transfer the same gamma power in the form of cesium-137 would have a shorter, smaller cavity, require only 8 inches of lead, weigh about 2.5 tons, and cost about \$6,000.

All handling cranes, etc., would have to be heavier to handle the shielded mixed fission products.

Efficient use of the gamma energy from the mixed fission products would require that the cylinders be spaced enough that most of the radiation would be absorbed before hitting another cylinder. In a water (or similar liquid) system this would require a spacing of about 2 feet between cylinder exteriors which would place the cylinder about 3 feet apart. If a radiation facility were to contain 216 cylinders spaced 3 feet on center, hexagonal, it would occupy about 1800 square feet of area and, assuming an overall height of 12 feet, about 22,000 cubic feet of contained volume.

In contrast, cobalt or cesium sources would not involve 1000 pounds of source for every 325 watts of gamma (average power over 18 years). Gamma absorption by sources would therefore be much less important and the same gamma use efficiency could probably be obtained by a facility one tenth the size--assuming dose rate and hydraulic limitations allowed it.

The shielding thickness required for a mixed fission product is about 25% greater than that required for a cobalt facility.

POSSIBLE USES, IRRADIATION

In comparison to pure isotopes, the use of mixed fission products as a source of irradiation presents two problems; the presence of low energy gammas (which must be eliminated if uniformity of dose is important) and the presence of high energy gammas (which increase the cost of shielding and transport but contribute little to total gamma power).

Quantitative calculation of the gamma spectrum delivered external to the cylinder, watts versus MeV, is beyond the scope of this report and is probably not justified until the waste solidification process to be used is determined. As an approximation, 60% of the gamma watts delivered external to the cylinder will consist of gammas attenuated by the shielding which will be distributed almost uniformly between about 0.15 MeV and the highest major peak (0.8 MeV in young wastes and 0.66 MeV in wastes over 10 years old). Thirty-two percent will consist of unattenuated primaries at 0.6, 0.66, and 0.8 with an average energy of 0.66. The remainder, 8%, will consist of high energy primaries and the high energy scatter from them.

Many of the possible uses for irradiation involve systems in which there are penalties for under-exposure or over-exposure or both. The gamma energy requirements vary widely with the individual case, sterilization of packaged bandages is most efficiently done by low energy irradiation (because they are thin in cross section) while preservation of large pieces of meat requires relatively high energy gammas if the center is to be sterilized without over-exposure of the surface. The calculation of individual cases is beyond the scope of this report.

Those wishing to calculate spectral distributions, dose rates, and integrated dose for individual cases may do so by assuming that, of the gamma watts available at the surface of the cylinder, 60% are evenly distributed between 0.15 and 0.65 MeV; 30% are at 0.66 MeV; and 10% are at 1.2 MeV. More precise calculation may be justified when actual cylinder dimensions, cylinder loading, and matrix compositions are fixed. For the present, however, the approximation suggested will furnish information on dose rate and integrated dose which is as accurate as the data now available.

The limitations in the use of gamma energy with this distribution should be appreciated. The dose rate will be approximately halved after passage through 10 inches of water or equivalent material. Its use to irradiate thick sections would therefore be limited although it might be made almost as good as ^{137}Cs (at some loss in gamma power efficiency) by use of a thin shield of lead or other heavy material with high attenuation for low energy gammas. In any case, it could never compete with cobalt-60 in any use requiring gamma energies greater than 1 MeV.

Some speciality uses involving the 2.18 MeV gamma from $^{144}\text{Ce-Pr}$ are conceivable but do not seem practical. One cylinder, weighing 1000 pounds, emitting initially 1000 watts of gamma power, and requiring an 18 ton cask, will furnish only 19 watts of 2.18 MeV gamma and it would be cheaper to obtain this amount of high energy gamma power by the use of a pure source.

Systems which can be well mixed or which can accept under- or over-exposure without major penalties can be handled more easily. Examples might include promotion of chemical reactions and irradiation of sewage. The approximate kilowatt hours

delivered each year by one cylinder of PWR waste have already been presented as a function of time in Table 9.

Knowing the gamma power emitted from the cylinder, one may calculate the irradiation which one cylinder can furnish. One kW-hr equals 36×10^{12} ergs and one rad (water) equals 100 ergs/gram or 378,540 ergs/gallon. A cylinder of 2 year waste could, therefore, expose a maximum of 1900 gallons per hour at 5×10^4 rad or 380 gallons per hour at 2.5×10^5 rad.

In an actual installation the cylinders would vary in age. The facility used as a basis of cost comparison, containing 216 cylinders varying in age from 2 to 20 years, would supply 70 kilowatts of gamma energy. If this were utilized at 75% efficiency to irradiate water, it could process 100,000 gallons per hour at 5×10^4 rad or 20,000 gallons per hour at 2.5×10^5 rad.

Such a facility could be used for commercial production of chemicals by process which include a chemical reaction requiring initiation by gamma irradiation. The annual production would depend upon the G value of the reaction (molecules produced per 100 electron volts absorbed). If 80% of the gamma watts delivered at the exterior of the cylinder were used to promote a reaction with a G value of 100 in a plant operating 90% of the year, the facility would produce 36,000 pound moles of product per year.

One potential use of irradiation which would require huge amounts of gamma power might be the use of radiation to sterilize the sewage from a major metropolitan area. This approach is being actively investigated and preliminary estimates of the dose necessary to kill bacteria and viruses have been made. See, for example, Touhill et al. (1968).

Some knowledge of its effect on noxious chemicals, and the biodegradability of the effluent stream is also available. An integrated dose of 2.5×10^5 rad appears to be necessary for a good kill of the microorganisms. Supplying this much radiation in a large, metropolitan sewage treatment plant, which might typically treat 1×10^9 gallons per day, would require 110,000 kW of gamma energy applied to the liquid. Supplying this amount of energy from mixed fission products is not practical. Even if one ignores the problems of initial inventory the maintenance of source would require 20,000 to 25,000 cylinders per year - roughly four times the production foreseen for the year 2000. Such a facility based upon mixed fission products or on fission product cesium is therefore out of the question for at least 50 years.

A similar irradiation facility based upon cobalt-60 is at least theoretically possible. At 25¢ per curie for cobalt-60 such a facility would cost \$2,000,000,000 for source and \$250,000,000 per year for source maintenance.

POSSIBLE USES, THERMAL ENERGY

Present uses of radioactive material as a source of thermal power are limited to applications which require a small amount of power in relatively inaccessible locations. The needs of this market are met by the use of relatively pure isotopes which are chosen to provide the optimum balance between cost, power, power density, and weight including the weight of the necessary shielding. Most of the isotopes used require relatively little shielding. In competing for this market, mixed fission products would have to bear the cost and inconvenience of the heavy shielding these materials require. While it is conceivable that mixed fission products could be used for some of these applications, the amount required would be small in comparison to the amount

available. As a result, expansion of present thermal uses is not expected to provide an important market for mixed fission products.

The best potential opportunity for the use of a major amount of mixed fission products as a source of thermal energy would appear to involve a large facility handling a substantial amount of material. Since the gamma spectrum of the material makes shielding, transport, and handling a major cost, this should be minimized as should the capital required.

It has previously been assumed that the solidified mixed fission products would require storage (to allow for decay of short-lived nuclides) for 1 or 2 years prior to industrial use or transport to permanent storage. If this is true, the most economical method of producing thermal power from the MFP would be to use the short-term storage facility as a source of heat. In doing this, both the rate of heat production and the amount of heat produced per unit of MFP would be maximized because the energy of the short-lived nuclides would be used. At the same time, capital cost would be minimized and handling cost would be eliminated. Whether the heat is used or not, any short-term storage cooling facility will have to shield and cool the MFP containers and the containers will have to be placed in the facility and later removed. Capital cost for a heat producing facility (over and above that required for a heat wasting short-term storage facility) would include the extra cost required by the distortion of the storage facility necessary to produce usable heat and the cost of any equipment required to use the heat. Extra handling, surveillance, and transportation costs would be minimal or nonexistent.

Value

The total energy liberated by one cylinder of mixed fission products while aging from 0.5 to 2.0 years is equivalent to about 408×10^6 Btu. Substantially all of this will be converted to heat while the cylinder is in the short-term storage facility and its value is the value of the fossil fuel necessary to furnish the same amount of heat. Prices for fossil fuel vary with location and size of user but recent prices for large amounts of fossil fuel, delivered, have ranged from 17¢ to 34¢ per million Btu [Hoyerton (1968)] with 25¢ as a national average [Shaw & Whitman (1968)]. Thermal efficiencies in the short-term storage facility would probably be less than the 80% obtained in commercial package boilers. Assuming 25¢ per million Btu for fossil fuel and a 67% thermal efficiency in the short-term storage facility, the heat released by one cylinder would be the same as that obtained from about \$100 worth of fossil fuel.

Plant Size and Cost Limitations

If one assumes a waste solidification plant which produces 1000 cylinders of mixed fission products per year (processing the wastes from 1000 tonne of BWR and 750 tonne of PWR fuel per year), the heat produced by the short-term storage facility would be equivalent to about 36,500 pounds of steam per hour. A package boiler to produce this much steam would cost about \$90,000 and require \$100,000 worth of fuel per year. If heat from the short-term storage facility is to be competitive, the (extra) capital required by the distortion of the facility to produce usable heat should not exceed \$90,000 except as it can be justified by the "free fuel" worth \$100,000 per year. If such an expenditure requires a 2 year write-off to be justified, the total extra capital required to produce usable heat should not

exceed \$290,000. These figures might be increased for a small scale user. If the competitive fuels were coal at \$25 per ton or natural gas at 5¢ per Therm. (100,000 Btu), the allowable cost might be doubled.

All of this assumes that there is a need for the heat at the site but this appears probable. If nothing else, steam will be required by both the chemical processing plant and the waste solidification facility.

Design Constraints

The primary functions of the short-term cooling facility will be to cool the cylinders of solidified waste enough to prevent damage from their own internally generated heat, provide distance and shielding enough to protect the surroundings from gamma radiation, and contain any fission products which might escape from an imperfect cylinder. Since the cylinders will have to be placed in and removed from the facility on a regular basis, it will probably be compartmentalized and have a number of loading ports. These ports, and the probability of their leaking will mean that the cylinders themselves will probably be stored at slightly less than atmospheric pressure.

If the heat produced in the short-term cooling facility is to be wasted, the cylinders will probably be cooled by being immersed in water boiling at or slightly below atmospheric pressure. The steam produced would be condensed for recycle, by an air or water cooled condenser.

If the facility is designed to produce saturated steam at or above atmospheric pressure, it would probably be necessary to remove the heat by transferring it from the cylinders to a high temperature fluid. The best material would probably be

water circulated in pipes under pressure although liquid metals or proprietary heat transfer materials might be used. Since the cylinders can be allowed a high surface temperature (425 °C) and have a high emissivity (0.85) it might be possible to design a facility which would allow transfer of all of the heat to the cooling coils by radiation. The distortion of the facility necessary to produce useful heat in this case would be the replacement of an immersion facility and condenser with a facility cooled by coils. This could have advantages. The facility would operate hot and dry and corrosion problems might be avoided. Contamination from an imperfect cylinder would not be distributed or carried by the cooling media.

An alternative approach might be to cool the cylinders with water boiling near atmospheric pressure, condense this for recycle by boiling water at still lower pressure, and raise the pressure of the secondary steam with a turbo compressor. This has never proven attractive in the United States; however, industrial installations do exist (e.g., in Norway where fuel is expensive and electric power is cheap).

Production of superheated steam could probably be handled in the same sort of high pressure coil system already discussed as a method of producing high pressure saturated steam. A detailed assessment of these possibilities is beyond the scope of this report, the amount of electric power which could be obtained would depend upon the pressure of the superheated steam and this would, in turn, depend on the design of the facility and the pressure drop in the coils.

An alternative to high pressure cooling coils might involve production of sub-atmospheric pressure superheated steam (e.g., 14 psia and 500 °F) with direct contact between the steam and the cylinders. Use of this steam would require a low pressure

steam turbine capable of remote operation (because the steam might be contaminated by a leaking cylinder). This could involve major design problems in the short-term cooling facility and in the turbine. If a facility were receiving 1000 cylinders per year and recovering 67% of the energy as heat, the electrical production would not exceed 1200 kW if the steam were produced at 14 psia and 500 °F or 2400 kW if produced at 100 psia and 800 °F.

In any case, if the heat is to be removed from the MFP containers by steam or air, the cooling fluid should enter the facility near the youngest wastes, which need the most cooling, and travel towards the oldest wastes. This is similar to "co-current" operation but it does not imply the poor efficiency normally associated with co-current heat exchangers. In contrast to the normal heat transfer problem, the rate at which heat is transferred from the heat source (the mixed fission products) is not determined by the temperature difference between the source and its surroundings. The reverse is the case. The heat is generated at a predetermined rate in each cylinder and the temperature of the cylinder adjusts itself to transfer the necessary heat to its surroundings.

Extraction of thermal energy other than in the short-term storage facility is technically possible, of course, but economics eliminate it. If all the energy liberated by one cylinder of mixed fission products were converted to thermal heat, the amount liberated between ages 0.5 years and 20.0 years (from reactor discharge) would be less than 1400×10^6 Btu (1126×10^6 Btu leaving the cylinder as heat plus 72,700 kW-hr leaving the cylinder as gamma energy which could be converted to an additional 248×10^6 Btu of heat). It would cost \$1030 to obtain

this cylinder and \$610 to dispose of it. Total cost = \$1640 per 1400×10^6 Btu. The same amount of heat delivered as fossil fuel would cost an industrial user less than \$1000, and the heat would be available immediately instead of being delivered over 18 years. The use of mixed fission products as sources of thermal energy (heat) is therefore eliminated by economics without even considering the (additional) capital cost required for the heat recovery facility. The writer can see no circumstances which could change this situation in the foreseeable future.

REFERENCES

J. F. Hoyerton. Scientific American, vol. 218, no. 2. 1968.

R. W. McKee, D. E. Deonigi and D. R. Haffner. Isotope Production Projections for U.S. Nuclear Power Reactors, BNWL-SA-1529. Presented at the American Chem. Soc. Meeting, San Francisco, Calif., April 1968; also, Isotope Production and Availability from Power Reactors, BNWL-716, Battelle-Northwest, Richland, Washington. July 1968.

M. Shaw and M. Whitman. Science and Technology, vol. 22. March 1968.

C. J. Touhill, E. C. Martin, M. P. Fujihara, D. E. Olesm, J. E. Stein and G. McDonnell. The Effects of Radiation on Chicago Metropolitan Sanitary District Municipal and Industrial Wastewaters, BNWL-SA-2106, Battelle-Northwest, Richland, Washington. 1968.

H. H. Van Tuyl. ISOGEN, A Computer Code for Radioisotope Generation Calculations, HW-83785. Available from Clearinghouse for Federal Scientific and Technical Information, Springfield, Virginia, 1964. (A current version of ISOGEN, modified to include power reactor cross sections, is available from the Argonne Code Center, Argonne National Laboratories, 9700 So. Cass Avenue, Argonne, Ill., 60439)

APPENDIX A

TABLES

TABLE 1. Nuclear Power Forecast

	Installed Capacity, gigawatts, elec.				Production, gigawatts, elec.	Total ¹³⁷ Cs at Discharge, kg	Tonne of Fuel		
	Total	PWR	BWR	FAST			PWR	BWR	FAST
1960	0.2	0.1	0.2		0.2				
61	0.4	0.2	0.2		0.4				
62	0.4	0.2	0.2		0.4	3			
63	0.6	0.2	0.3		0.5	16			
64	0.6	0.2	0.3		0.5	23			
65	0.6	0.2	0.3		0.5	14			
66	0.9	0.5	0.4		0.8	20			
67	1.8	1.4	0.4		1.6	25			
68	2.9	1.5	1.4		2.4	32	15	16	
69	5.5	2.8	2.7		4.7	71	32	37	
70	11.7	5.9	5.8		10.0	64	28	34	
71	21.7	10.9	10.9		18.5	157	69	83	
72	32.0	16.0	16.0		27.2	366	161	193	
73	42.9	21.5	21.4		36.5	684	301	361	
74	54.2	27.2	27.0		46.1	1080	476	571	
75	66.2	39.2	33.0		56.3	1507	664	796	
76	79.2	39.7	39.5		67.3	1886	830	997	
77	93.7	46.9	46.8		79.6	2276	1002	1203	
78	110.2	55.2	55.0		93.6	2710	1193	1432	
79	128.9	64.5	64.4		109.5	3209	1413	1696	
80	150.0	74.0	74.0	2.0	127.4	3723	1618	1941	13
81	173.7	83.9	83.8	6.0	147.6	4436	1886	2263	40
82	200.2	93.1	93.0	14.0	170.0	5215	2156	2563	96
83	229.4	103.2	103.1	23.0	194.8	6159	2440	2885	163
84	261.3	114.2	114.1	33.0	221.8	6968	2681	3217	233
85	295.5	125.8	125.7	44.0	250.6	7867	2948	3537	309
86	332.1	138.1	138.0	56.0	281.2	8684	3179	3815	387
87	370.6	150.8	150.8	69.0	313.2	9845	3527	4232	484
88	411.2	164.1	164.1	83.0	346.7	10954	3850	4620	584
89	454.1	177.6	177.5	99.0	381.9	12313	4240	5088	709
90	500.0	191.5	191.5	117.0	419.3	13581	4581	5497	840

TABLE 2. *Availability of (PWR and BWR) Mixed Fission Product Cylinders (Wastes Aged 2 Years from Reactor Discharge)*

<u>Year</u>	<u>Cylinders/Year</u>
1970	18
1975	381
1980	1511
1985	3070
1990	4877

TABLE 3. *Amount of Wastes in PWR and BWR Fuels Aged Two Years from Reactor Discharge*

	<u>Grams Per Tonne of Fuel</u>	
	<u>PWR</u>	<u>BWR</u>
¹⁵⁵ Eu	1.837	1.483
¹⁵⁴ Eu	27.80	21.26
¹⁴⁷ Eu	83.19	71.23
¹⁴⁴ Eu	0.0022	0.0015
¹⁴⁴ Ce	52.09	34.58
¹³⁷ Cs	1087	901.4
¹³⁴ Cs	63.75	43.31
^{125m} Cs	0.057	0.046
¹²⁵ Sb	4.362	3.494
^{110m} Ag	0.0072	0.0055
¹⁰⁶ Ru	30.37	23.49
⁹⁵ Nb	0.0284	0.0189
⁹⁵ Zr	0.0243	0.0162
⁹¹ Y	0.0059	0.0045
⁹⁰ Y	0.1272	0.0983
⁹⁰ Sr	486.2	386.6
⁸⁹ Sr	0.0010	0.0007

TABLE 4. Gamma Spectrum of PWR Mixed Fission Products

*Basis: Waste Fission Products from 1 tonne PWR Fuel
at 30,000 MWd/tonne and 32.5 MW/tonne*

		Age, yr								
		0.5	0.75	1.0	1.5	2.0	3.0	5.0	10.0	20.0
MeV	Nuclide	Watts of Gamma Power								
<0.13	144Ce	15.5	12.7	10.5	7.4	5.3	3.0	1.5	0.9	0.5
0.134		53.9	43.2	34.6	22.2	14.2	5.9	1.0	0.01	---
		143	38.7	17.5	11.0	9.4	7.1	4.2	1.2	0.3
0.512	106Ru	180.4	151.7	127.5	90.2	63.7	31.9	8.0	0.3	---
0.563	134Cs	37.7	34.7	32.0	27.1	23.0	16.5	8.5	1.6	0.06
0.569	134Cs	62.8	57.8	53.2	45.1	38.3	27.1	14.2	2.7	0.1
	134Cs	7.3	6.5	6.1	5.4	4.8	3.9	2.2	1.1	0.4
0.605		467.3	430.4	396.2	335.9	284.8	204.7	105.8	20.3	0.8
0.622		106Ru	117.6	98.9	83.1	58.8	41.6	20.8	5.2	0.2
	137Cs	9.4	4.5	3.3	2.5	2.2	1.6	1.0	0.3	0.02
0.662		322.8	321.0	319.1	315.4	311.8	304.7	290.9	259.2	205.7
		1.1	1.0	1.0	0.8	0.8	0.7	0.5	0.3	0.2
0.696	144Pr	38.1	30.5	24.5	15.7	10.1	4.1	0.7	0.008	---
0.697-0.796	134Cs	2335.0	922.8	357.8	55.6	11.8	4.5	3.9	3.2	2.1
0.797		533.9	491.7	452.8	383.8	325.4	233.9	120.9	23.2	0.9
0.802		56.9	52.4	48.2	40.9	34.9	24.9	12.9	2.5	0.09
0.803-0.999	106Ru	12.2	9.4	8.1	6.4	5.3	4.0	2.9	2.1	1.4
1.00-1.06		42.8	36.5	32.3	25.9	21.3	15.3	9.6	5.7	3.4
1.14		7.8	6.6	5.5	3.9	2.8	1.4	0.4	0.01	---
1.168	134Cs	18.4	17.0	15.6	13.2	11.2	8.1	4.2	0.8	---
1.249	154Eu	0.4	0.3	0.3	0.3	0.3	0.3	0.3	0.2	0.2
1.278	154Eu	12.4	12.3	12.1	11.9	11.6	11.1	10.2	8.2	5.3
1.31	106Ru	1.1	0.95	0.8	0.6	0.4	0.2	0.05	0.002	---
1.365	134Cs	37.7	34.7	31.9	27.1	22.0	16.5	8.5	1.6	---
	144Pr	0.7	0.5	0.4	0.3	0.2	0.1	0.01	0.00007	---
1.49		16.1	12.9	10.3	6.6	4.3	1.7	0.3	0.003	---
		1.4	1.3	1.1	0.8	0.7	0.4	0.3	0.2	0.1
1.56-1.565	106Ru+	3.5	2.9	2.5	1.7	1.2	0.6	0.2	0.005	---
1.6	154Eu	0.8	0.8	0.8	0.7	0.7	0.7	0.6	0.5	0.3
1.76	106Ru	1.2	1.0	0.9	0.6	0.4	0.2	0.05	0.002	---
1.93	106Ru	1.0	0.8	0.7	0.5	0.4	0.2	0.04	0.001	---
2.13	106Ru	1.8	1.5	1.3	0.9	0.6	0.3	0.08	0.003	---
2.186	144Pr	59.5	47.6	38.1	24.5	15.7	6.5	1.1	0.01	---
2.37	106Ru	3.3	2.7	2.3	1.6	1.2	0.6	0.1	0.005	---
2.44	106Ru	0.3	0.3	0.2	0.2	0.1	0.1	0.02	0.0005	---
2.63	106Ru	0.2	0.2	0.2	0.1	0.1	0.04	0.01	0.0003	---
2.88	106Ru	0.2	0.2	0.1	0.1	0.1	0.04	0.009	0.0003	---

TABLE 5. Gamma Spectrum of BWR Mixed Fission Products

Basis: Waste Fission Products from 1 tonne BWR Fuel
at 25,000 MWd/tonne and 20.3 MW/tonne

		Age, yr							
		0.5	1.0	1.5	2.0	3.0	5.0	10.0	20.0
MeV	Nuclide	Watts of Gamma Power							
<0.13	144Ce	10.59	7.23	5.11	3.73		1.09	0.67	0.41
0.134		35.79	22.96	14.73	9.44		0.66	0.008	---
		103.05	13.04	8.26	7.08		3.17	0.94	0.19
0.512	106Ru	139.48	98.59	69.71	49.31		6.16	0.19	---
0.563	134Cs	25.62	21.72	18.42	15.62		5.80	1.11	0.04
0.569	134Cs	42.65	36.16	30.66	26.00		9.66	1.85	0.07
	134Cs	5.82	4.83	4.29	3.25		2.05	0.88	0.34
0.605		317.4	269.14	228.18	193.48		71.87	13.80	0.51
0.622	106Ru	90.92	64.27	45.44	32.14		4.02	0.13	---
	137Cs	7.25	2.59	2.03	1.73		0.76	0.34	0.02
0.662		267.70	264.60	261.56	258.56		241.25	214.93	170.58
	144Pr	0.85	0.74	0.66	0.60		0.39	0.24	0.14
0.696		25.31	16.23	10.41	6.68		0.47	0.006	---
	134Cs	1552.67	238.36	37.41	8.31		3.00	2.42	1.57
0.797		362.68	307.52	260.72	221.07		82.11	15.77	0.58
0.802	134Cs	38.64	32.77	27.78	23.55		8.75	1.68	0.06
		9.33	6.28	4.93	4.09		2.19	1.60	1.03
1.00-1.06		32.13	24.16	19.37	15.90		6.39	4.32	2.62
1.14	106Ru	6.06	4.28	3.03	2.14		0.27	0.008	---
1.168	134Cs	12.51	10.60	8.99	7.62		2.83	0.54	0.02
1.249	154Eu	0.27	0.26	0.26	0.25		0.22	0.18	0.12
1.278	154Eu	9.48	9.28	9.08	8.88		7.80	6.28	4.07
1.31	106Ru	0.87	0.62	0.43	0.31		0.04	0.001	---
1.365	134Cs	25.58	21.69	18.39	15.59		5.80	1.11	0.04
	144Pr	0.47	0.29	0.18	0.12		0.007	---	---
1.49		10.69	6.85	4.40	2.82		0.20	0.002	---
	106Ru+	1.11	0.83	0.64	0.50		0.21	0.14	0.09
1.56-1.565		2.71	1.91	1.35	0.96		0.12	0.004	---
1.6	154Eu	0.59	0.58	0.57	0.56		0.49	0.39	0.03
1.76	106Ru	0.94	0.66	0.47	0.33		0.04	0.001	---
1.93	106Ru	0.77	0.54	0.39	0.27		0.03	0.001	---
2.13	106Ru	1.42	1.00	0.71	0.50		0.06	0.002	---
2.186	144Pr	39.48	25.31	16.24	10.4		0.73	0.009	---
2.37	106Ru	2.52	1.78	1.26	0.89		0.11	0.003	---
2.44	106Ru	0.26	0.18	0.13	0.09		0.01	---	---
2.63	106Ru	0.17	0.12	0.09	0.06		0.008	---	---
2.88	106Ru	0.15	0.11	0.08	0.05		0.007	---	---

TABLE 6. *Major Sources of Gamma Power (Watts)*

<u>Nuclide</u>	<u>2 Years</u>		<u>10 Years</u>	
	<u>PWR</u>	<u>BWR</u>	<u>PWR</u>	<u>BWR</u>
^{134}Cs	58.4%	54.5%	15.9%	13.5%
^{137}Cs	24.3	27.7	77.0	79.7
^{106}Ru	9.6	10.2		
^{154}Eu	2.4	2.5	6.4	6.1
^{144}Pr	2.4	2.2		
^{144}Ce	1.4	1.3		
^{125}Sb	0.9	1.0		

TABLE 7. *Effect of Self-Shielding and Cylinder Wall Shielding*

<u>Initial γ MeV</u>	<u>Fraction of Initial Power</u>	<u>Primary Attenuation</u>	<u>Buildup</u>	
0 - 0.5	(0.0225)	(~ 0.05)	(~ 4)	= ~ 0.0045
0.5 - 0.8	(0.90)	(0.187)	(2.8)	= 0.47124
0.8 - 2.5	(0.0775)	(0.3)	(2.5)	= <u>0.05813</u>
				0.533

TABLE 8. *Power Level (Watts) of PWR and BWR Cylinders*

Age, Years Since Reactor Discharge	PWR			BWR		
	Thermal	Gamma	Total	Thermal	Gamma	Total
0.5	17883	3675	21558	17631	3389	21020
0.75	13452	2298	15750			
1.0	10768	1696	12464	10661	1609	12276
1.5	7526	1228	8754	7451	1183	8634
2.0	5573	1020	6593	5531	989	6520
3.0	3429	766	4195			
5.0	1932	493	2425	2034	498	2531
10.0	1302	267	1570	1441	286	1727
20.0	1045	176	1221	1199	194	1393

Note: These data include the effect of self-shielding and cylinder wall shielding, i.e., thermal watts are $\alpha + \beta + 47\%$ of γ ; γ watts are 53% of those emitted by the mixed fission products.

TABLE 9. *Energy from One Cylinder of PWR Mixed Fission Products (MFP)*

Time Period		$\alpha+\beta$ Emitted By MFP, kW-hr	γ Emitted By MFP, kW-hr	Heat From Cylinder, Btu	γ 's From Cylinder, kW-hr
From	To				
0.5	2.0	100460	40730	408 $\times 10^6$	21590
2.0	3.0	28080	14040	118 $\times 10^6$	7440
3.0	4.0	17730	11010	78.2 $\times 10^6$	5835
4	5	12400	8960	56.7 $\times 10^6$	4750
5	6	9930	7460	45.9 $\times 10^6$	3950
6	7	8480	6360	39.1 $\times 10^6$	3370
7	8	7665	5600	35.1 $\times 10^6$	2970
8	9	7150	5040	32.5 $\times 10^6$	2670
9	10	6960	6400	31.1 $\times 10^6$	2440
10	11	6890	4200	30.3 $\times 10^6$	2225
11	12	6825	3850	29.5 $\times 10^6$	2040
12	13	6770	3600	28.9 $\times 10^6$	1910
13	14	6720	3420	28.4 $\times 10^6$	1810
14	15	6680	3240	28.0 $\times 10^6$	1720
15	16	6630	3130	27.6 $\times 10^6$	1660
16	17	6590	3060	27.4 $\times 10^6$	1620
17	18	6560	3000	27.2 $\times 10^6$	1590
18	19	6530	2950	27.0 $\times 10^6$	1560
19	20	6500	2920	26.9 $\times 10^6$	1550
Totals				1126 $\times 10^6$	72700
0.5	2.0			408 $\times 10^6$	21590
2.0	20.0			718 $\times 10^6$	51110
0.5	3.0			526 $\times 10^6$	29030
3.0	20.0			600 $\times 10^6$	43670
0.5	5.0			661 $\times 10^6$	39620
5.0	20.0			465 $\times 10^6$	33090

TABLE 10. *Cost of 70 Kilowatt Gamma Source*

<u>Source Type</u>	<u>Basis</u>	<u>Initial</u> + <u>Annual</u>
Mixed Fission Product	Shipping Costs Only	\$ 87,000 \$ 10,000/year (1st 15 Years) \$ 20,000/year (later)
Cesium-137	5¢/curie delivered	\$1,200,000 + \$ 27,000/year
	10¢/curie delivered	\$2,400,000 + \$ 54,000/year
Cobalt-60	25¢/curie delivered	\$1,425,000 + \$177,000/year
	50¢/curie delivered	\$2,850,000 + \$353,000/year

APPENDIX B

FIGURES

FIGURE 1. *Installed Capacity - Nuclear Electrical Generation*

FIGURE 2. *Gamma Spectra for PWR Mixed Fission Products
(2 yr from reactor discharge)*

FIGURE 2. Gamma Spectra for PWR Mixed Fission Products
(2 yr from reactor discharge)

FIGURE 3. *Gamma Spectra for PWR Mixed Fission Products
(3 yr from reactor discharge)*

FIGURE 4. *Gamma Spectra for PWR Mixed Fission Products
(5 yr from reactor discharge)*

FIGURE 5. *Gamma Spectra for PWR Mixed Fission Products
(10 yr from reactor discharge)*

FIGURE 6. *Gamma Spectra for PWR Mixed Fission Products
(20 yr from reactor discharge)*

FIGURE 7. Comparison of Gamma Spectra from PWR and BWR Wastes

FIGURE 8. Comparison of Mixed Fission Products and ^{137}Cs Gamma Watts

DISTRIBUTIONNo. of
CopiesOFFSITE

1 AEC Chicago Patent Group
 G. H. Lee

194 AEC Division of Technical Information Extension

ONSITE-HANFORD

✓1 AEC Chicago Patent Group
 R. K. Sharp (Richland)

✓2 AEC RDT Site Representative
 P. G. Holsted

3 AEC Richland Operations Office
 B. J. Melton *Lab. + Univ. Div.*
 C. L. Robinson
 B. A. Ryan

✓4 Atlantic Richfield Hanford Company
 M. K. Harmon
 R. E. Isaacson
 R. E. Tomlinson
 P. W. Smith

✓3 Battelle Memorial Institute

✓42 Battelle-Northwest

J. M. Atwood
A. G. Blasewitz
D. W. Bolme (25)
R. E. Burns
J. E. Hansen
R. S. Kemper
A. M. Platt
K. J. Schneider
R. J. Thompson
H. H. Van Tuyl
Legal

✓ Technical Information (5)
Technical Publications (2)